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		Examiner Name	H. WILKINS III
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Firm	Sean P. O'Hanlon Swidler Berlin LLP		
Signature			
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Date	Dec. 14, 2005	Reg. No.	47,252

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of: M. Dahlbäck *et al.*

Application No.: 09/857,606

Group Art Unit: 1742

Filed: August 2, 2001

Examiner: H. Wilkins III

For: ZIRCONIUM BASED ALLOY AND COMPONENT IN A NUCLEAR ENERGY
PLANT

APPEAL BRIEF

Mail Stop Appeal Brief - Patents
Commissioner for Patents
PO Box 1450
Alexandria, Virginia 22313-1450

Sir:

This appeal is from the final Office Action mailed June 21, 2005 finally rejecting all of the pending claims. A Notice of Appeal was timely filed on September 21, 2005, along with a Pre-Appeal Brief Request for Review. A Notice of Panel Decision from Pre-Appeal Brief Review was issued on November 14, 2005. This Brief is being filed under the provisions of 37 CFR § 1.192.

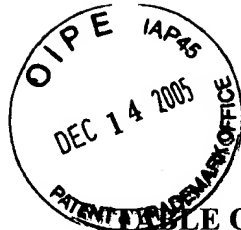


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REAL PARTY IN INTEREST

Westinghouse Electric Sweden AB, assignee of the instant application by virtue of the documents filed with the Assignment Division on January 25, 2005, is the real party in interest.

RELATED APPEALS AND INTERFERENCES

None.

STATUS OF CLAIMS

Claims 13, 22, 23, and 35-40 are pending, and claims 1-12, 14-21, and 24-34 are canceled. Claims 13, 22, 23, and 35-40 were rejected in the final Office Action dated June 21, 2005. The Appellants appeal the final rejection of claims 13, 22, 23, and 35-40.

STATUS OF AMENDMENTS

No amendments were filed subsequent to final rejection. All amendments filed by the Appellant have been duly entered by the Examiner.

SUMMARY OF CLAIMED SUBJECT MATTER

The present invention as claimed relates to a cladding tube for nuclear fuel. The cladding tube is made of a zirconium-based alloy, suitable for use in a corrosive environment where it is subjected to increased radiation. *See, e.g.*, Written Description at page 1, lines 10-15¹. The composition is formulated such that the alloy also exhibits excellent physical and mechanical properties. *Id.* at page 3, line 33 to page 4, line 4. To achieve these benefits, the alloy as now claimed contains: 0.65-1.6 weight percent niobium (Nb) to provide strength and to enhance corrosion resistance (*Id.* at page 7, lines 25-32; page 3, lines 7-9); 0.3-0.6 weight percent iron (Fe) to enhance corrosion resistance (*Id.* at page 7, lines 25-32; page 1, line 26 to page 2, line 3); 0.65-0.85 weight percent tin (Sn) to enhance the corrosion resistance (*Id.* at page 4, lines 13-30); the balance of the alloy being zirconium (Zr) (*Id.* at page 7, lines 25-32).

Of course, the alloy may contain impurities. *Id.* at page 9, lines 17-18. Small amounts of silicon and oxygen may be present. *Id.* at page 9, lines 18-21. The exact amount of these impurities will necessarily vary due to manufacturing. The alloy may thus include up to 1600 ppm oxygen and up to 120 ppm silicon. Typically expected amounts of silicon are 50-120 ppm, and typically expected amounts of oxygen are 500-1600 ppm. *Id.* at page 9, lines 15-30.

The cladding tube is designed for use in a nuclear power plant. *Id.* at page 6, lines 5-6. The inventive alloy is ideal for such a component due to its ability to withstand heightened corrosion effects of fast neutron radiation that is present in such an environment, especially around the reactor core. *Id.* at page 6, lines 5-31.

According to one embodiment, at least a part of the cladding tube inner circumference has a layer of a material which is more ductile than the inventive alloy. *Id.* at page 6, lines 33-

¹ Citations are to the PCT publication (WO 00/36170).

36. This may help ensure that the cladding tube is less sensitive to the direct contact with the fuel and that the risk of cracking is reduced. *Id.* at page 6, line 36 to page 7, line 7. To this end, the ductile layer include a zirconium-based alloy with a total content of alloying materials which does not exceed 0.5 percentage by weight. *Id.* at page 7, lines 7-9.

GROUND OF REJECTION TO BE REVIEWED ON APPEAL

The grounds of rejection to be reviewed on appeal are:

1. Whether claims 13, 22, and 23 are unpatentable under 35 U.S.C. § 112, first paragraph.
2. Whether claims 13, 22, 23, and 35-37 are unpatentable under 35 U.S.C. § 102 over U.S. Patent No. 5,023,048 to Mardon *et al.* (Mardon) as supported by U.S. Patent Nos. 5,790,623 to Van Swam (Van Swam) and 5,254,308 to Garde *et al.* (Garde '308).
3. Whether claims 38-40 are unpatentable under 35 U.S.C. § 103 over Mardon modified by U.S. Patent No. 5,211,774 to Garde *et al.* (Garde '774).

ARGUMENT

Rejections under 35 U.S.C. § 112, first paragraph

The Examiner rejected claims 13, 22, and 23, alleging that the claims contain subject matter that was not adequately described in the specification. Specifically, the Examiner stated that “[t]he specification only provides support for the range of O being 500-1600 ppm.”

Claim 13 recites “... the alloy having a quality and impurity level, including up to 1600 ppm O” This recitation is supported in the specification. As expressly stated, “small amounts of Si and O *may* exist in the alloy.” Written Description at page 9, lines 20-21 (emphasis added). Clearly, this means that small amounts of oxygen may exist in the alloy or there may be no oxygen in the alloy. The Appellants expressly state a range of oxygen that typically may be expected in the alloy - 500-1600 ppm. *Id.* at page 9, lines 29-30. This, however, is not the only amount of oxygen that may be present. As previously established, *small amounts* of Si and O may exist in the alloy. If 500-1600 ppm qualifies as a “small amount” of oxygen, surely 0-500 ppm must also qualify as a small amount of oxygen. Furthermore, because the exemplary range of oxygen is described as being *typical*, it cannot be read to be the *only* range possible. *See Id.* at page 9, lines 21-22. Finally, it is nonsensical to think that 500-1600 ppm is the only amount of oxygen qualifying as an impurity level. *See Id.* at page 9, lines 17-18, 20-21.

Thus, “up to 1600 ppm O” is disclosed and supported in the written description, and the Examiner’s rejections under 35 U.S.C. § 112, first paragraph, of claims 13, 22, and 23 are improper and must be withdrawn.

Rejection under 35 U.S.C. § 102 over Mardon as supported by Van Swam Garde '308

Claims 13 and 35

Mardon appears to disclose a so-called duplex tube. Such a duplex tube comprises a main tubular element provided with an outer surface layer. It is clear from the abstract that this layer only constitutes 10-25% of the total thickness of the wall. The concept “duplex tube” is also explained in, for example, column 1, lines 49-62. Mardon thus suggests a particular duplex tube. The main portion of this tube is made of a zirconium alloy of conventional type (see, for example, column 2 lines 55-68). These conventional alloys are very different from the inventive alloy of the present invention. Consequently, Mardon does not teach the present invention.

In contrast to Mardon, the present invention discloses and claims a cladding tube *made of* the defined alloy. *Cf.* Written Description at page 6, lines 28-29; page 7, lines 22-23. According to one embodiment, the cladding tube may be provided with an inner protective layer. As is known in art, such a layer is often called a liner, and such a layer is rather thin compared to the thickness of the alloy from which the cladding tube is made. It is thus clear from the wording of the claims, in view of the description and in accordance with how the terms are used in the art, that the present invention claims a cladding tube for which the main part (or the entirety) is formed by the novel alloy defined in the independent claims. For this reason alone it is clear that the present invention is new and inventive and patentable over Mardon.

Furthermore, Mardon does not teach that its thin outer layer should constitute the base material for the tubular element. On the contrary, as explained above, this layer is only used as an external protective layer. The composition of the alloy in this protective layer is given in examples 1-3 in column 3. Example 1 defines an Fe range that does not at all overlap with the Fe range of the alloy of present invention. Example 3 includes V but not Nb, and is therefore not in accordance with the alloy of the present invention. Example 2 includes an Fe content that is

within the range defined in the present independent claims. However, example 2 teaches a lower Sn content, that only coincides with the range defined in the present independent claims at the exact value 0.65. Furthermore, example 2 in the cited document teaches a Nb content that is essentially lower than according to the present invention. The Nb content only coincides with the range defined in the present independent claims at the exact value 0.65. Furthermore, Mardon example 2 only mentions ranges for each of the elements of the alloy; it does not teach any concrete example of how to choose the exact amounts of the different elements. A person skilled in the art, who would like to fabricate the tubular element according to Mardon, has, by necessity, to choose some particular values for the different alloying elements. There is no indication in Mardon that a person skilled in the art would choose exactly the upper limit (0.65) for Sn and at the same time select the very highest upper limit for the Nb content (0.65). This means that Mardon does not suggest the alloy defined in the independent claims of the present application. On the contrary, even the alloy according to Mardon example 2 teaches away from the alloy defined in the present independent claims, since Mardon teaches a lower range for the Sn content and a lower range for the Nb content. Mardon therefore does not suggest an alloy according to the present invention even in connection with the outer protective layer described therein. Moreover, Mardon does not even suggest such an alloy for the main portion of the tubular element.

The Examiner has alleged that the Appellants are attempting to ascribe a novel meaning to the term *cladding tube*, and further states that “[a] cladding tube is any tube shaped object which surrounds a cylindrical object.” June 21, 2005 final Office Action at section 8b. The Appellants respectfully traverse the Examiner’s statements. The examiner equates the thinner protective surface layer in the duplex tube of Mardon with a cladding tube. This external layer

itself is not a cladding tube as the term is known or used in the art. Instead, it constitutes just a thinner protective surface layer. *See* Mardon at column 1, lines 49-63; column 2, lines 21-27. The main part of the tube according to Mardon is made of a conventional alloy. Such an alloy is very different from the alloy of which the cladding tube according to the present invention is made. Consequently, Mardon does not disclose a cladding tube as defined in the claims. Furthermore, as explained above, not even the thin external layer with which the tube according to Mardon is provided defines the same alloy as the alloy of which the cladding tube according to the present invention is made.

Thus, the Examiner's rejections of claims 13 and 35 are improper and must be withdrawn.

Claims 22 and 36

Claims 22 and 36 are dependent claims. Consequently, the subject matter of these claims is novel for the above reasons regarding claims 13 and 35. Further regarding claims 22 and 36, the Examiner has stated that Mardon teaches the use of a conventional zirconium alloy for the inner layer of its tube, and relies on Van Swam to define the conventional zirconium alloy. This reliance upon Van Swam is improper. In rejecting the independent claims from which claims 22 and 36 depend, the Examiner relied upon the disclosure of Mardon to define the composition of the inner layer of its tube. The Examiner then attempts to re-define the very same composition to be something different when addressing the dependent claims. The Examiner cannot assign such contrasting definitions to a single term. In doing so, the Examiner has removed the definition of the composition of the cladding tube, and thus the Examiner has not addressed each of the claim elements.

Moreover, the Examiner cannot alter the express teaching of Mardon of the inner layer composition (*see* Mardon at column 2, lines 55-68 - "[t]he inner tubular layer is made from a

zirconium alloy of conventional type *Such* a zirconium-base alloy, designated Zircaloy 4, includes” (emphasis added)) without providing proper motivation for doing so, despite the Examiner’s improper designation of the rejection as being an anticipation-type rejection rather than an obviousness-type rejection.

Moreover, as explained above, the present invention claims a cladding tube made from a certain alloy. This cladding tube is, according to the embodiment defined in claims 22 and 36, provided with an inner layer of a more ductile material. The Examiner equates this inner layer according to the invention with the main component, made of a standard alloy, in the duplex tube according to Mardon. This is improper and not in accordance with how the terms are used in the art.

Furthermore, in order to arrive at this conclusion, the Examiner starts from the improper premise, as explained above, that the external thinner duplex layer of Mardon corresponds to the cladding tube according to the present invention as defined in claims 13 and 35.

Nevertheless, the Examiner states that the inner layer of Mardon is more ductile than the alloy of the outer layer. This does not seem to be correct. It is explicitly stated in Mardon, that the “inner layer” is of a conventional alloy such as Zircaloy-4 (*see* column 2, lines 55-68) or alternatively Zircaloy-2 or a conventional zirconium-base alloy with about 1% niobium (*see* column 3, lines 40-49; column 4, lines 23-28). As explained above, Mardon concerns a so-called duplex tube. As far as the inventors of the present invention are aware, in a duplex tube normally the alloy of the main, inner component is less ductile than the alloy of the outer duplex layer. There is no indication in Mardon that the opposite would be the case for the tube disclosed therein.

The Examiner argues that the main component of Mardon should be Zr with 0.4 Fe in accordance with the teaching of Van Swam. The Examiner refers to column 7, lines 45-49 in Van Swam. *See* June 21, 2005 final Office Action at section 4. However, the layer that it is here being referred to is an innermost layer 127 in a rather unusual three-component tube. *See* Mardon at column 7, lines 6-13. The “inner layer” in Mardon, on the other hand, is the main component in a duplex tube. It is explicitly stated that this component is of a conventional kind such as Zircaloy-4 or, alternatively, Zircaloy-2 or Zr-1% Nb. *See also* claims 5, 6, and 7 in Mardon. It is therefore improper, and contrary to the teaching of Mardon, to rely on the teaching concerning the inner layer in the three-component tube according to Van Swam as a support for the ductility concerning the conventional layer in Mardon.

Thus, the Examiner’s rejections of claims 22 and 36 are improper and must be withdrawn.

Claims 23 and 37

The Examiner has not addressed the recitations of claims 23 and 37, and as such these claims have not been rejected.

Rejections under 35 U.S.C. § 103 over Mardon modified by Garde ‘774

Claim 38

The statements made above regarding the Examiner’s improper use of the Mardon reference to claims 13 and 35 are equally applicable to claim 38, and are thus incorporated herein.

Concerning claim 38, the Examiner argues that the subject matter of this claim is obvious in view of Mardon in combination with Garde. June 21, 2005 final Office Action at section 6.

In this context, the Examiner still refers to the outer surface layer in Mardon. As explained above, the alloy in this layer is not relevant to the present invention.

Nevertheless, the Examiner argues that it would be obvious to add 50-120 ppm Si to the alloy in this outer duplex layer of Mardon in view of Garde. *Id.*

Garde teaches a particular alloy that is said to have advantageous properties concerning corrosion resistance, ductility, and reduced hydrogen absorption. *See* Garde at the Abstract. Garde explicitly teaches that the niobium content should be at the most 0.6%, typically 0.3%. *See, for example*, column 3 lines 31-39. The amount of niobium is thus clearly lower than in the alloy according to the present invention.

If a person skilled in the art would at all be interested in combining the teaching of Mardon with that of Garde, this person would make sure that the niobium content is lower than 0.6% in accordance with the teaching of Garde. *See Id.* The person skilled in the art would therefore not arrive at an alloy as defined in the pending claim 38. On the contrary, Garde clearly supports the non-obviousness of the present invention, since this document teaches that a person skilled in the art should *not* select the highest value of niobium in the range specified in Mardon.

Moreover, Garde teaches that the alloy should include 0.1-0.4% Cr. *See, for example*, Garde column 3, lines 24-29; Table 1. However, Mardon explains that the outer layer shall not contain any chromium at all. *See, for example*, Mardon at the table in column 3; column 3, lines 21-22. Consequently, it would not be obvious to apply the teaching of Garde to Mardon. Because Garde concerns a completely different alloy than the alloy in the outer layer of Mardon, it would not be obvious to select one individual element (silicon) out of context in Garde and to use this amount out of context in the outer layer of Mardon.

It should also be noted that Garde is completely based on the idea of modifying the contents of alloying elements in traditional alloys such as Zircaloy-4. *See, for example*, Garde at column 2, lines 52-62. If the teaching of Garde were to be combined with the teaching of Mardon, it would therefore be in order to modify the alloy of the base material of Mardon, as this material typically can be Zircaloy-4. *See, for example*, Mardon at column 2, lines 55-68. However, such a combination would not lead to the present invention.

Furthermore, the Examiner's statement that Mardon anticipates claim 38 is erroneous and in direct contrast with the Examiner's previous admission that Mardon does not disclose or suggest all of the elements of claim 38. *See* the June 21, 2005 final Office Action at sections 7 and 6, respectively. The Examiner's refusal to consider the declaration of Magnus Limbäck, which was submitted with the Response to the January 28, 2005 non-final Office Action, is improper. The Examiner must consider the declaration.

The declaration, together with the previously filed declaration, is in fact highly relevant to the non-obviousness of the present invention. From the declarations it is clear that the present invention provides unexpected and advantageous results.

Thus, the Examiner's rejections of claim 38 is improper and must be withdrawn.

Claim 39

Claim 39 is identical to claims 22 and 36, but for its dependence from a different independent claim. The statements made above regarding claims 22 and 36, therefore, are equally applicable to claim 39 and are thus incorporated herein. Thus, the rejection of claim 39 is improper and must be withdrawn.

Claim 40

Claim 40 is identical to claims 23 and 37, but for its dependence from a different independent claim. The statements made above regarding claims 23 and 37, therefore, are

equally applicable to claim 40 and are thus incorporated herein. Thus, the rejection of claim 40 is improper and must be withdrawn.

CLAIMS APPENDIX

1 13. A cladding tube for nuclear fuel, the cladding tube being made of a zirconium-based
2 alloy suitable for use in a corrosive environment where it is subjected to increased radiation, the
3 alloy having a quality and impurity level, including up to 1600 ppm O and up to 120 ppm Si,
4 suitable for use in nuclear reactors, the alloy consisting essentially of:

5 0.65-1.6 percent by weight Nb;
6 0.3-0.6 percent by weight Fe;
7 0.65-0.85 percent by weight Sn; and
8 the balance being Zr.

1 22. The cladding tube according to claim 13, wherein at least a part of an inner circumference
2 of the cladding tube is provided with a layer of a material that is more ductile than the alloy.

1 23. The cladding tube according to claim 22, wherein the layer comprises a zirconium-based
2 alloy having a total content of alloying elements that does not exceed 0.5 percent by weight.

1 35. A cladding tube for nuclear fuel, the cladding tube being made of a zirconium-based
2 alloy suitable for use in a corrosive environment where it is subjected to increased radiation, the
3 alloy having a quality and impurity level, including, optionally, 500-1600 ppm O and, optionally,
4 50-120 ppm Si, suitable for use in nuclear reactors, the alloy consisting essentially of:

5 0.65-1.6 percent by weight Nb;
6 0.3-0.6 percent by weight Fe;
7 0.65-0.85 percent by weight Sn; and
8 the balance being Zr.

1 36. The cladding tube according to claim 35, wherein at least a part of an inner circumference
2 of the cladding tube is provided with a layer of a material that is more ductile than the alloy.

1 37. The cladding tube according to claim 36, wherein the layer comprises a zirconium-based
2 alloy having a total content of alloying elements that does not exceed 0.5 percent by weight.

1 38. A cladding tube for nuclear fuel, the cladding tube being made of a zirconium-based
2 alloy suitable for use in a corrosive environment where it is subjected to increased radiation, the
3 alloy having a quality and impurity level, including 500-1600 ppm O and 50-120 ppm Si,
4 suitable for use in nuclear reactors, the alloy consisting essentially of:

5 0.65-1.6 percent by weight Nb;

6 0.3-0.6 percent by weight Fe;

7 0.65-0.85 percent by weight Sn; and

8 the balance being Zr.

1 39. The cladding tube according to claim 38, wherein at least a part of an inner circumference
2 of the cladding tube is provided with a layer of a material that is more ductile than the alloy.

1 40. The cladding tube according to claim 39, wherein the layer comprises a zirconium-based
2 alloy having a total content of alloying elements that does not exceed 0.5 percent by weight.

EVIDENCE APPENDIX

1. A copy of the declaration of Magnus Limbäck, which was submitted with the Response to the January 28, 2005 non-final Office Action, is attached hereto as Exhibit A.
2. A copy of the declaration of Magnus Limbäck, which was submitted with the Response to the December 24, 2003 non-final Office Action, is attached hereto as Exhibit B.

RELATED PROCEEDINGS APPENDIX

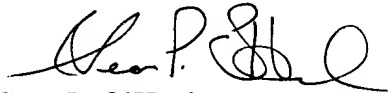
None.

CONCLUSION

In view of the foregoing arguments, the Appellants respectfully request reconsideration and withdrawal of the claim rejections, and that the application be passed to issuance. Failing that, the Appellants respectfully request the Board to overrule the Examiner's rejections, based on the explanations presented above, and to pass this application to issuance.

The Commissioner is hereby authorized to charge the appeal brief fee set forth in 37 CFR 41.20(b)(2) and any insufficiency or credit any overpayment associated with this application to Swidler Berlin LLP Deposit Account No. 19-5127 (order no. 19378.0011).

Respectfully Submitted,



Sean P. O'Hanlon
Reg. No. 47,252

Dated: December 14, 2005

Swidler Berlin LLP
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DECLARATION

Re: US patent application 09/857606

I, Magnus Limbäck, M.Sc. in Engineering Physics, employed at Westinghouse Electric Sweden AB and there responsible for BWR fuel material, make the following declaration.

I.

I have previously made a declaration, signed by me on 22 June 2004, in this case.

I have now noticed that the declaration contains typing errors in connection with Figure 3. These errors are that the same alloy (Zr1.5Nb0.5Sn0.3Fe) has been entered twice in the legend to the right of the figure and twice in the rectangles pointing to the figure itself.

I here below, on the next page, enclose a corrected Figure 3. The places where the figure has been corrected are marked by arrows pointing to the corrected entries.

When further below in this declaration Figure 3 is mentioned, it is being referred to the corrected Figure 3. When Figures 1 and 2 are mentioned, it is being referred to these Figures in the previous declaration.

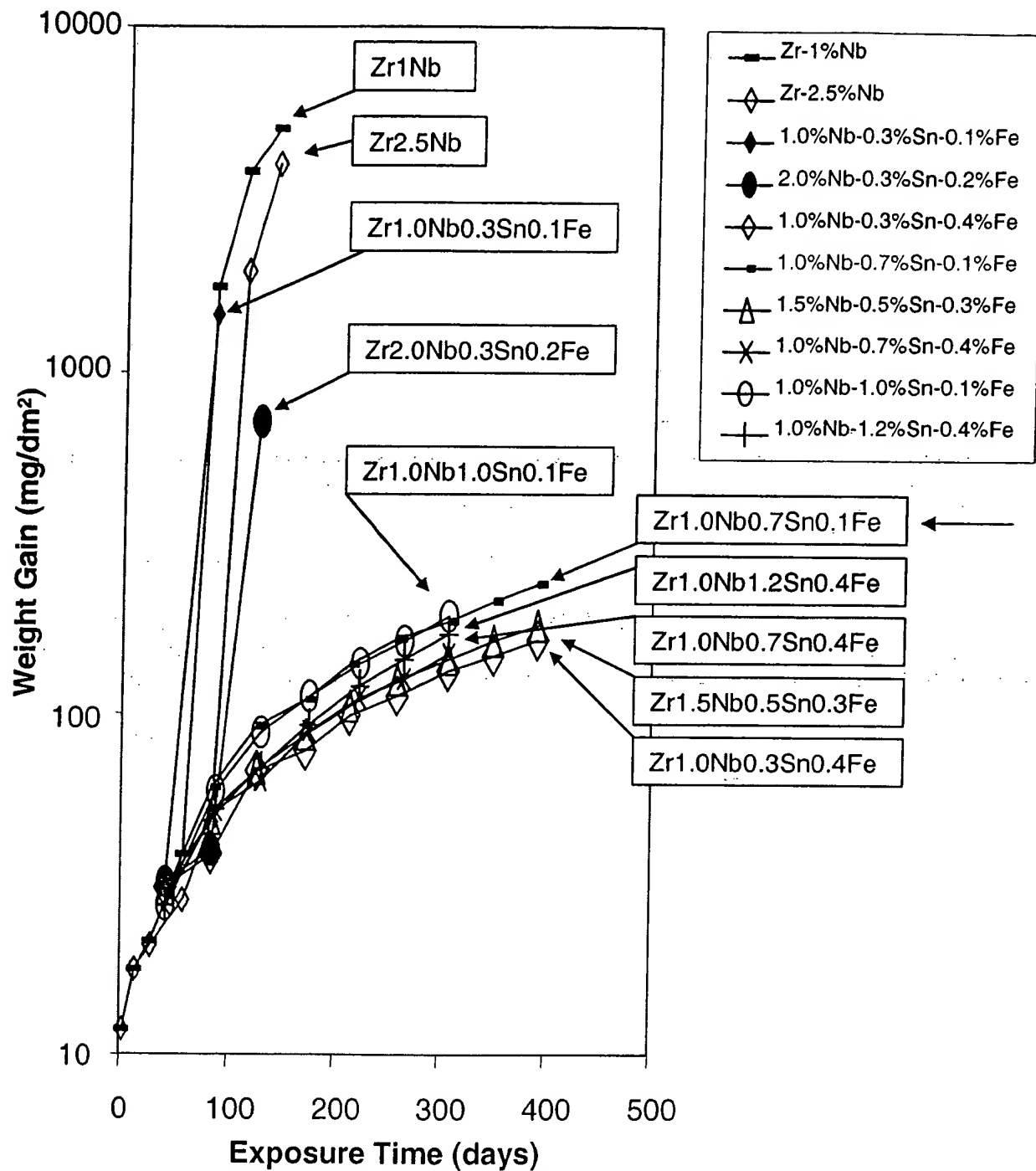


Figure 3: Long term corrosion test of coupons in 70 ppm lithiated water at 633 K.

II.

In addition to the to the statements made above, and in the previous declaration, I hereby add the following.

The results from the 633 K pure water (Fig. 2) and the 700 K steam (Fig. 1) tests indicate that the Sn concentration should be as low as possible within the studied interval to optimize the corrosion resistance in these environments. On the other hand, Fig. 3, which presents the results from the 633 K 70 ppm lithiated water tests, shows two classes of materials, one that is sensitive to Li-induced acceleration of the corrosion rate and one that is almost immune against this type of acceleration. For the inventors of the product described in the current patent application it has been important to assure that the newly developed material will belong to the second improved group.

It is, thus, of special interest to compare the results for four of the materials included in Fig. 3, $\text{Zr}_{1.0}\text{Nb}_{0.3}\text{Sn}_{0.1}\text{Fe}$, $\text{Zr}_{1.0}\text{Nb}_{0.7}\text{Sn}_{0.1}\text{Fe}$, $\text{Zr}_{1.0}\text{Nb}_{0.3}\text{Sn}_{0.4}\text{Fe}$ and $\text{Zr}_{1.0}\text{Nb}_{0.7}\text{Sn}_{0.4}\text{Fe}$. The first of these four materials experiences Li-induced acceleration of the corrosion rate, while the latter three have very good resistance against such acceleration. It is concluded that the difference in performance between these alloys is due to the difference in chemical composition. The alloy with poor corrosion resistance has low Sn (0.3 wt%) and low Fe (0.1 wt%) contents, while the alloys with very good resistance have higher Sn (0.7 wt%) and/or higher Fe (0.4 wt%) concentrations. The data, consequently, shows that the corrosion resistance is improved by increasing the Sn concentration from 0.3 to 0.7 wt% and by increasing the Fe content from 0.1 to 0.4 wt%.

The effect of Sn and Fe concentration on the corrosion performance is further elucidated by plotting subsets of the data presented in Fig. 3. Figure A, consequently, depicts the weight gain measured after 84 days as a function of Sn concentration for $\text{Zr}_{1.0}\text{Nb}_x\text{Sn}_{0.1}\text{Fe}$ type materials. This graph shows the significant improvement in corrosion resistance that is gained by increasing the Sn content from 0.3 to 0.7 wt%. Figure B, furthermore, shows the beneficial effect of increasing the Fe concentration in $\text{Zr}_{1.0}\text{Nb}_{0.7}\text{Sn}_x\text{Fe}$ type materials that is observed after 307 days.

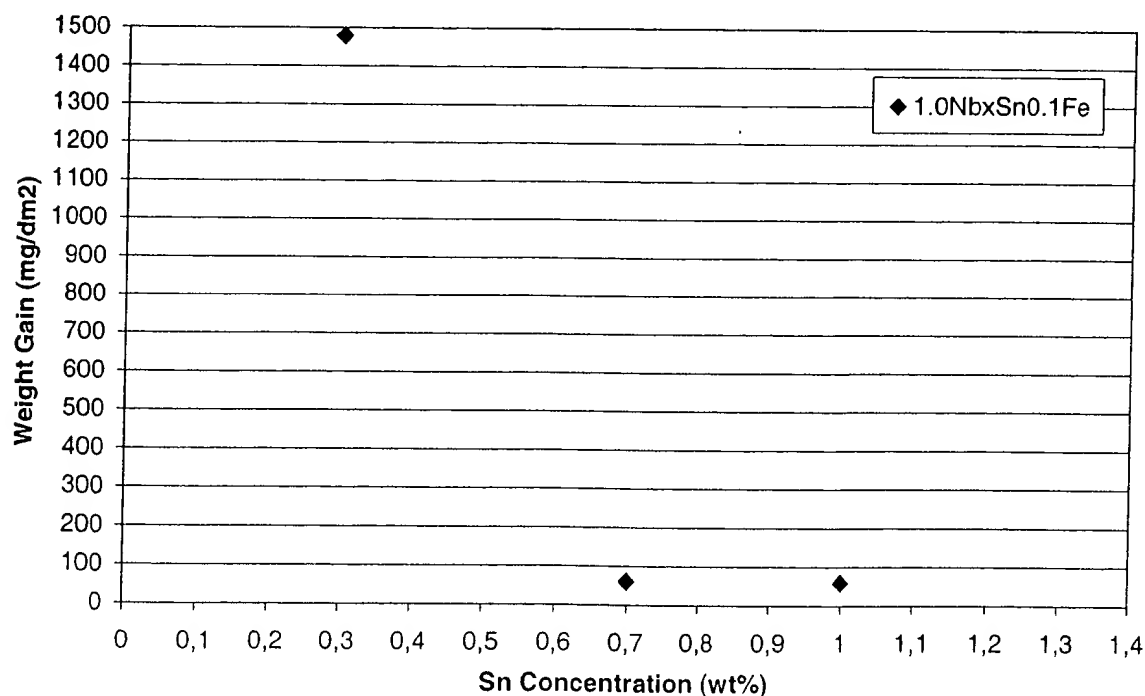


Figure A: Weight gain as a function of Sn content for $\text{Zr}_{1.0}\text{Nb}_x\text{Sn}_{0.1}\text{Fe}$ type materials after 84 days exposure at 633 K in 70 ppm lithiated water.

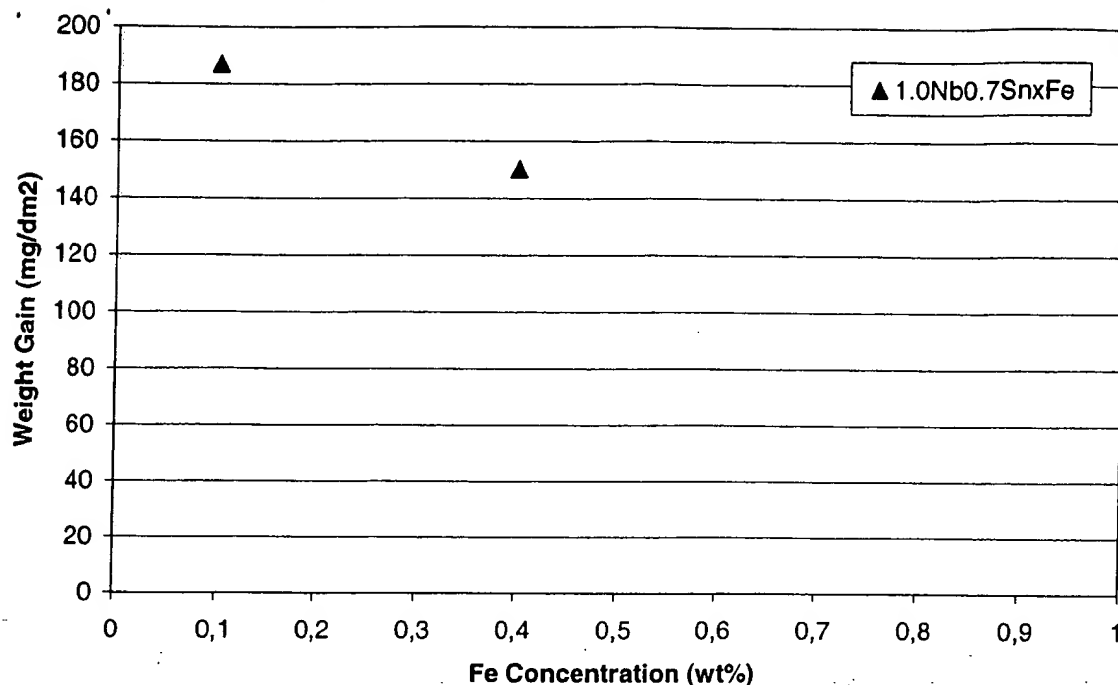


Figure B: Weight gain as a function of Fe content for Zr1.0Nb0.7SnxFe type materials after 307 days exposure at 633 K in 70 ppm lithiated water.

The goal of the current development is an alloy that is robust and has very good corrosion resistance not only in autoclaves but also, and more importantly, in various types of in-reactor environments. The inventors have applied a redundant approach where the two improving factors, i.e., increased Fe and Sn concentrations, are combined. The inventors have, consequently, chosen an alloy with >0.65 wt% Sn in combination with >0.30 wt% Fe.

In addition to the test results presented earlier a corrosion test has also been made in steam at 415°C (688 K). The results are presented in Fig. C and show that decreasing the Sn content from 1.14 to 0.95 drastically improves the corrosion resistance. The data, moreover, show that decreasing the Sn content to 0.8 wt% further improves the corrosion resistance, but that the material with 0.65 wt% Sn has about the same corrosion resistance as the material with 0.8 wt% Sn. These observations are all valid for materials with 0.32-0.35 wt% Fe. When comparing the two materials with 0.65-0.67 wt% Sn it can be observed that the material with the higher Fe content (0.35 wt%) has somewhat better corrosion resistance than the material with lower Fe concentration (0.10 wt%).

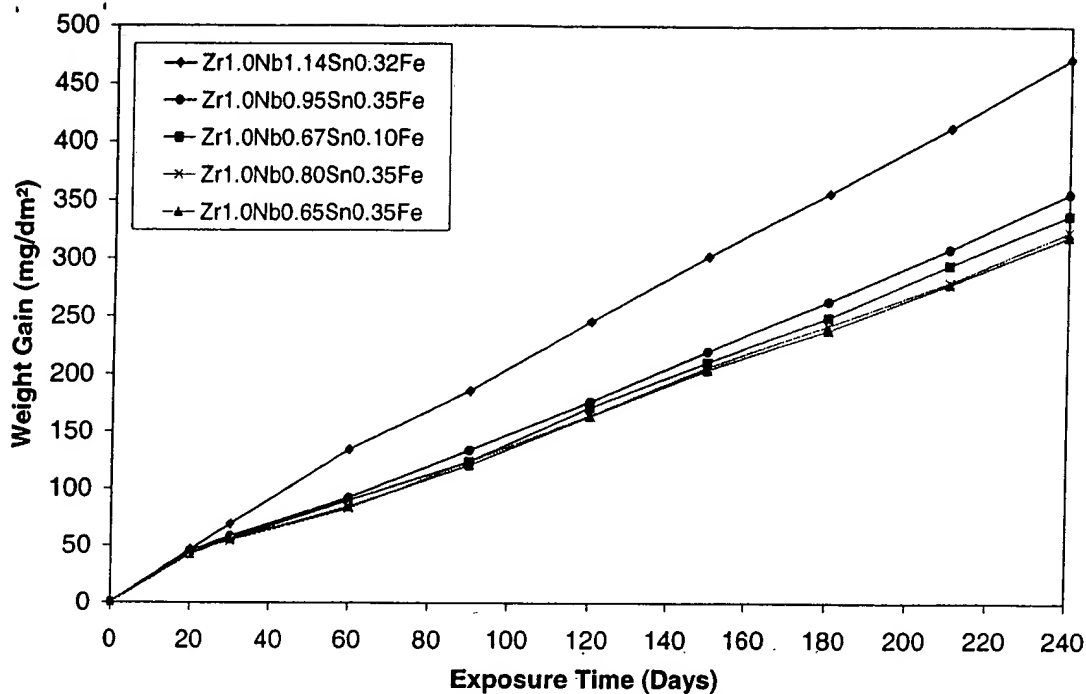


Figure C: Long term autoclave test results of coupons in steam at 688 K.

Based on the results from the 633 K 70 ppm lithiated water, 633 K pure water and 700 K steam tests presented earlier along with the 688 K steam test presented here, the inventors conclude that the Sn concentration should be within a rather narrow region, 0.65-0.85 wt%. The inventors, further, conclude that the Fe concentration should be 0.3-0.6 wt%, where the lower limit is chosen to secure good corrosion resistance while the upper limit is chosen to minimize potential fabrication issues, i.e., secure a good production yield.

I declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Västerås, Sweden, 26 April 2005

Magnus Limbäck

DECLARATION

Re: US patent application 09/857606

I, Magnus Limbäck, M.Sc. in Engineering Physics, employed at Westinghouse Electric Sweden AB and there responsible for BWR fuel material, make the following declaration concerning tests made of alloys for use in nuclear energy plants.

The tests were performed by Westinghouse Electric Corporation, USA, in cooperation with Westinghouse Electric Sweden AB.

The corrosion performance of alloys of the type $Zr_xNb_ySn_zFe$ (Zr alloy with x, y and z wt% Nb, Sn and Fe, respectively.) in different environments has very recently been presented by one of the inventors behind the current patent application along with five co-authors [1]. The studied material types are presented in Table 1.

The corrosion performance was evaluated by autoclave tests that were conducted in a manner consistent with ASTM Practice for Aqueous Corrosion Testing of Samples of Zirconium and Zirconium Alloys (G2). Exposures in 633 K water were performed at saturation pressure while exposures at 700 K steam were performed at 10.3 MPa. Testing at 633 K included both pure water and lithiated water containing 70 ppm Li that was added as lithium hydroxide. The autoclave tests were periodically interrupted to obtain corrosion weight gains of the specimens. Testing included three specimens per alloy, and the reported corrosion data represent the average specimen weight gains. The corrosion test results in steam at 700 K, pure water at 633 K and 70 ppm lithiated water at 633K are depicted in Figs. 1-3, respectively.

The corrosion behavior in steam at 700 K and pure water at 633 K was quantified by calculating the post transition corrosion rate for each alloy composition by performing a linear least squares fit of the data. The rates were calculated over the time interval of 140 to 469 days for 633 K water and 61 to 251 days for 700 K steam. Also included for comparisons in the corrosion evaluations are results from two Zr-Nb binary alloy samples that were previously reported [2].

The effect of chemistry on corrosion behavior was examined by assessing the impact of the chemistry variations on corrosion rates. Corrosion rates were plotted as a function of individual alloying elements in order to identify potential relationships between alloy chemistry and corrosion behavior. Figure 4 shows the relationship between normalized corrosion rates in 633 K water and 700 K steam as a function of three chemistry variables (Nb, Sn, and Fe). The rates were normalized relative to those of an alloy with a nominal chemistry of Zr-1Nb-1Sn-0.1Fe.

Figure 4a shows the impact of tin on the corrosion rate. Even with variations of other alloying elements, the impact of tin on the corrosion rate in the two environments is apparent. In both pure water and steam environments, the corrosion rate decreases with decreasing tin content. The rate of change in the autoclave tests is about a 5% decrease in corrosion rate for each 0.1% decrease in tin. Figures 4b and 4c show the relative corrosion rates plotted as functions of the niobium and iron contents, respectively. Figure 4b shows that there is no significant correlation with niobium variations. Likewise Figure 4c shows that the effects of variation in iron content on corrosion rates in these environments are secondary in comparison to the tin effects.

Figure 3 is a plot of corrosion weight gains of the various alloys in 70 ppm lithiated water at 633K. In the lithiated water environment, the corrosion rate is only slightly affected by the tin content until it is below a threshold tin value. This suggests that a minimum critical tin content is needed to prevent accelerated corrosion in the lithiated water environment. While the tin content was not studied as an entirely independent variable, non-linear multiple regression analysis was utilized to evaluate the impact of the main alloying elements on the uniform corrosion rate in lithiated water. The result of this analysis indicates that tin is clearly an important factor in the resistance to accelerated corrosion in high lithium environments.

In addition to the beneficial impact of tin on the uniform corrosion rate in the lithiated environment, there is also indication that iron may provide a similar effect. A comparison of two alloys, Zr1.0Nb0.3Sn0.1Fe and Zr1.0Nb0.3Sn0.4Fe, shows iron also contribute to the delay of the accelerated corrosion in high lithium environment. Figure 5 is a plot of relative corrosion rates in 70 ppm lithiated water as a function of the combined iron and tin levels. The results indicate that a combination tin and iron in the alloy provides resistance to accelerated corrosion from 70 ppm lithiated water autoclave tests. It was suspected that the individual effects of tin and iron on lithium accelerated corrosion are not totally equivalent but the initial comparison as shown in Figure 5 indicates the effects of both tin and iron.

In-reactor corrosion is a complex phenomenon and the 70 ppm Li environment is used to simulate the Li accelerated corrosion that might occur in the upper spans of high power fuel assemblies, which may experience concentrated lithium conditions in the oxide due to sub-cooled boiling at the cladding surface. A second possible Li concentration mechanism is boiling in crud on the rod surface. With recent utilities' desire to operate at higher coolant lithium concentrations to control coolant pH and reduce crud formation, lithium corrosion resistant alloys are an important part of robust fuel designs.

It is, finally, concluded that the results from the various tests and analyses strongly support the invention described in the current patent application. The proposed chemical composition provides a material with very good corrosion resistance in all of the studied environments. In addition, the presence of tin at a well optimized level, as the one put forward in the patent application, helps to maintain good mechanical strength, primarily creep and tensile strengths.

Table 1: Nominal alloy chemistry.

Alloy ID	Alloy Elements Composition (wt%)		
	Nb	Sn	Fe
Group 1 (variable tin)			
1-1	1.0	1.3	0.1
1-2	1.0	1.0	0.1
1-3	1.0	0.7	0.1
1-4	1.0	0.3	0.1
Group 2 (high iron, variable tin)			
2-1	1.0	1.2	0.4
2-2	1.0	0.7	0.4
2-3	1.0	0.3	0.4
Group 3 (high niobium, high iron, low tin)			
3-1	1.5	0.5	0.3
3-2	2.0	0.3	0.2

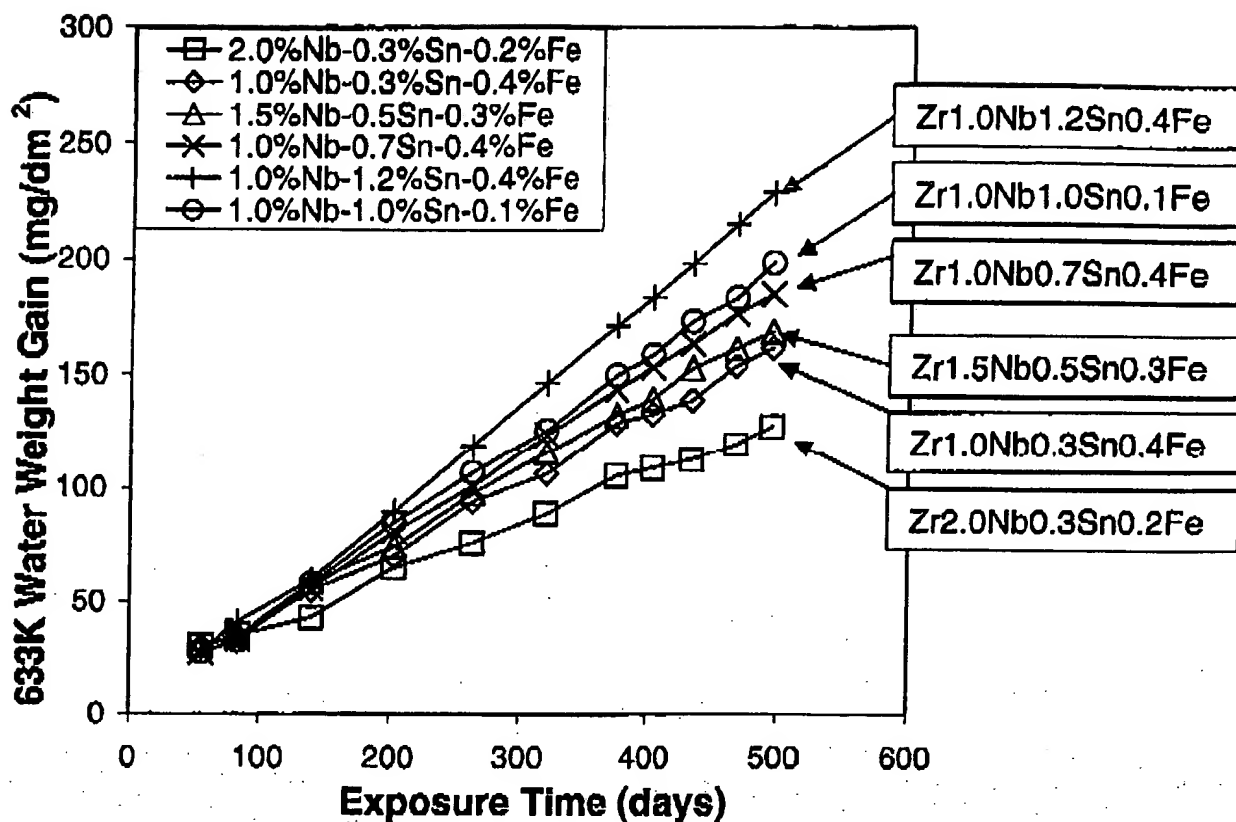


Figure 1: Long term autoclave test results of coupons in steam at 700 K.

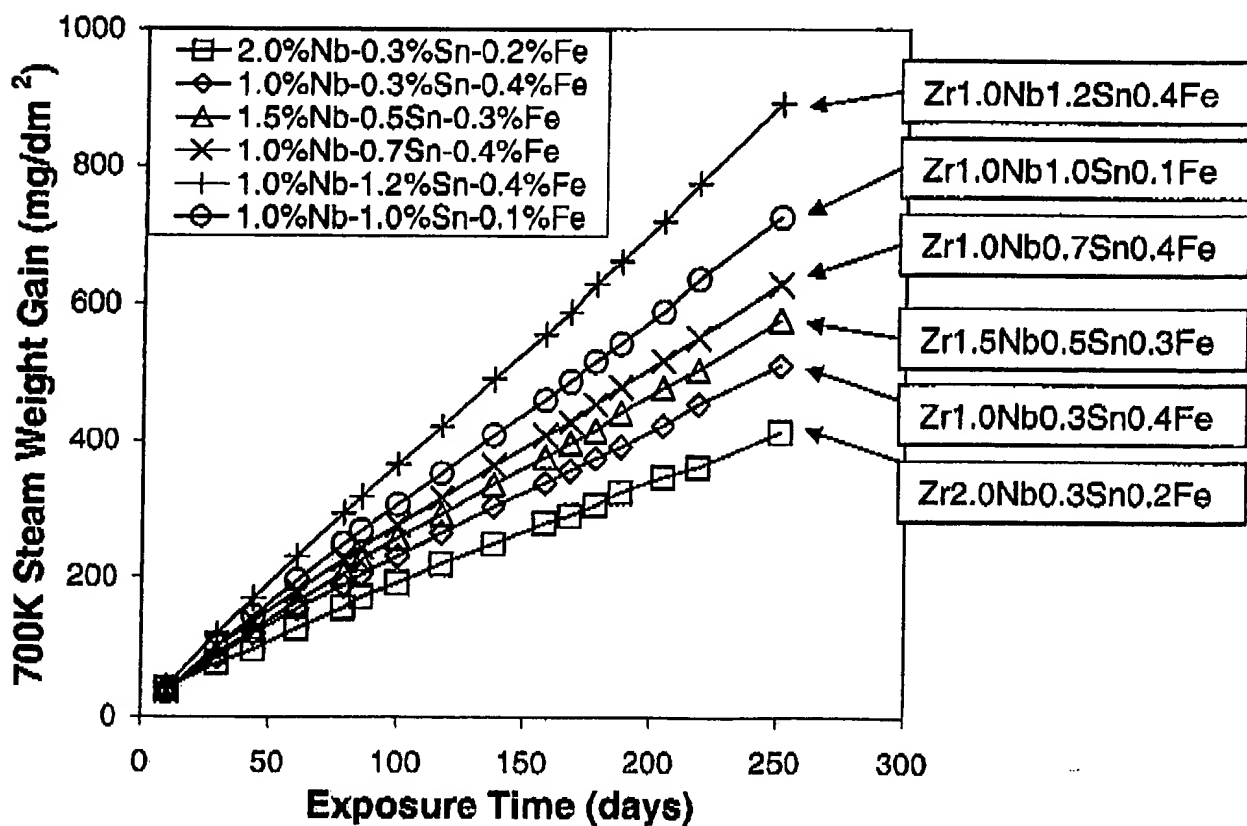


Figure 2: Long term autoclave test results of coupons in pure water at 633 K.

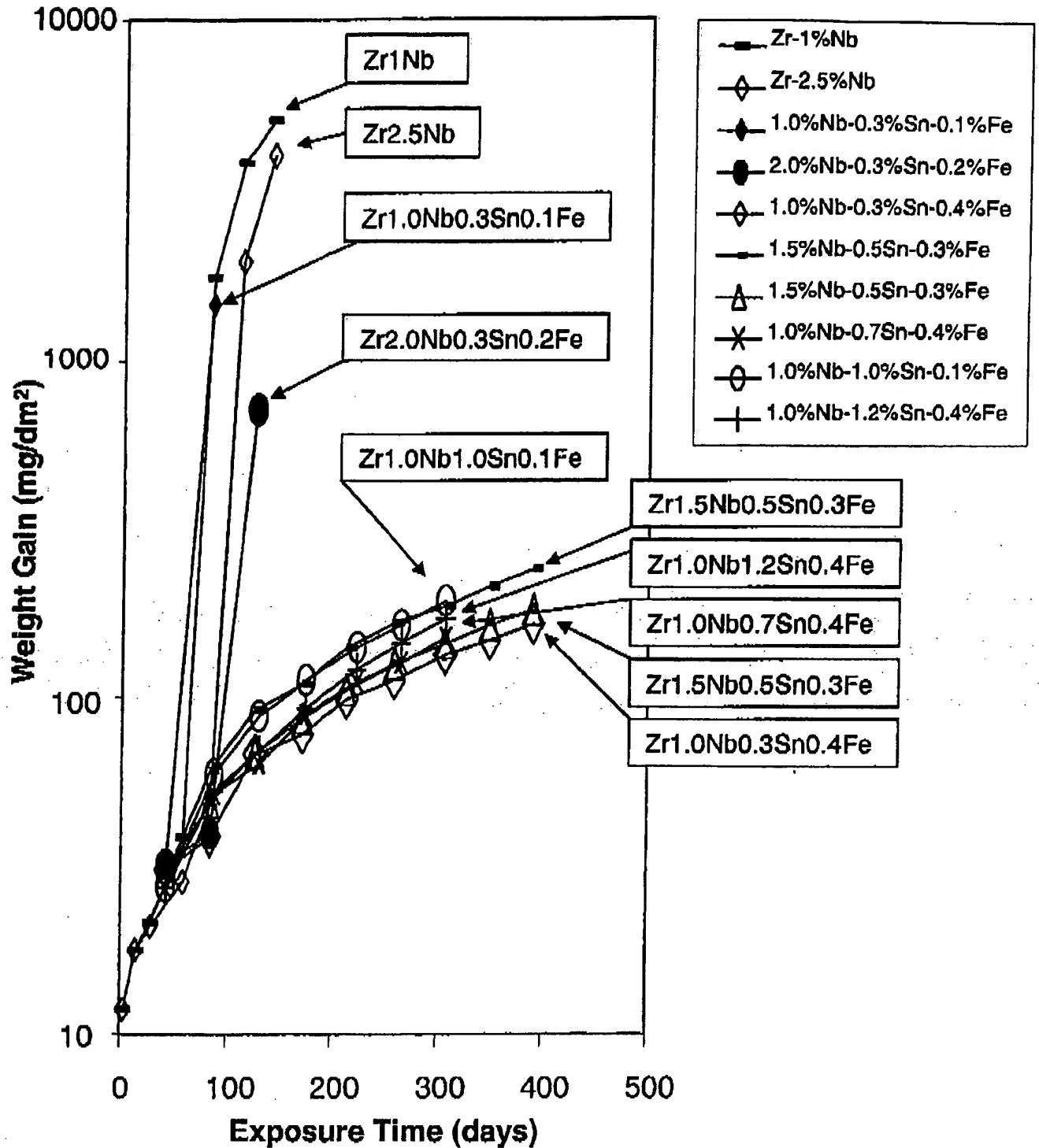
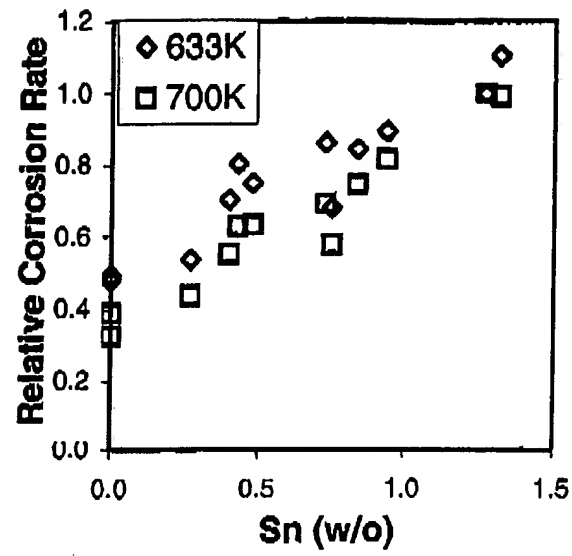
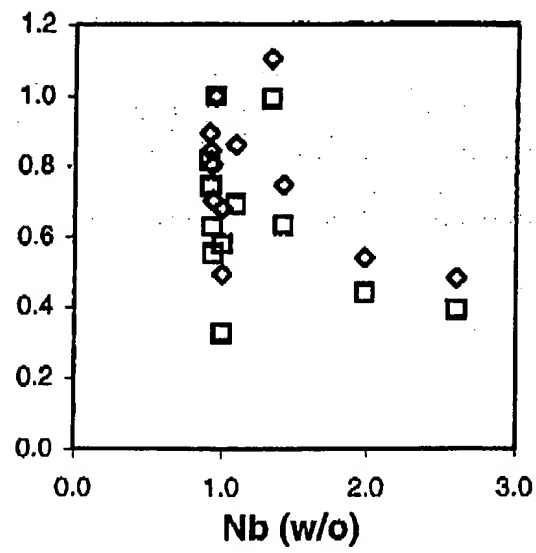


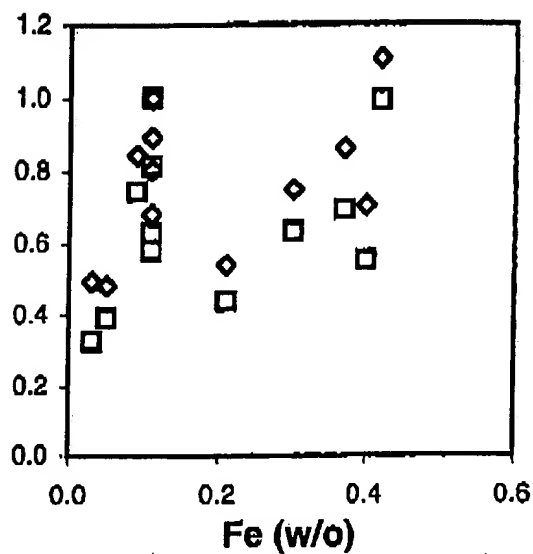
Figure 3: Long term autoclave test results of coupons in 70 ppm lithiated water at 633 K.



a)



b)



c)

Figure 4: Variation in normalized (to 1%Sn-1%Nb-0.1%Fe) corrosion rate as a function of chemistry variable: a) tin, b) niobium, and c) iron.

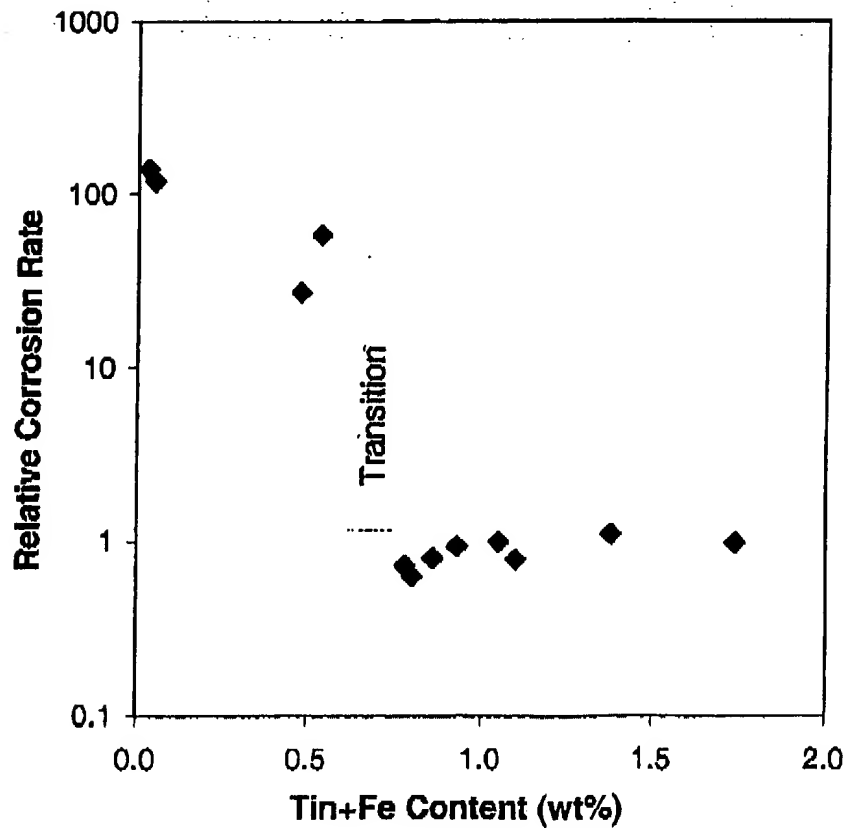


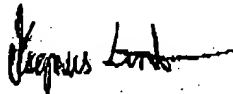
Figure 5: Impact of tin and iron content on corrosion rate in 633K-70 ppm lithiated water environment.

References

- [1] H.K. Yuch, R.L. Kesterson, R.J. Comstock, D.J. Colburn, M. Dahlbäck and L. Hallstadius, "Improved ZIRLO™ Cladding Performance through Chemistry and Process Modifications", Paper presented at the ASTM 14th International Symposium on Zirconium in the Nuclear Industry, Stockholm, Sweden, June 13-17, 2004.
- [2] R.J. Comstock, G. Schoenberger and G.P. Sabol, "Influence of Processing Variables and Alloy Chemistry on the Corrosion Behavior of ZIRLO Nuclear Fuel Cladding", ASTM STP 1295, 1996, P710.

I declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Västerås, Sweden, 22 June 2004



Magnus Limbäck